REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	10/13/95	Final 5/92 - 8/95

4. TITLE AND SUBTITLE

Two-Photon Cooperative Cascade Superfluorescence

5. FUNDING NUMBERS

19951101 102

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8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

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12a. DISTRIBUTION/AVAILABILITY STATEMENT

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12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

We present a study of the time development of an ensemble of three-level atoms which have been put in a coherent superposition of their ground and excited states (of the same parity) by the application of a short two-photon resonant laser pulse. We worked with Cs vapor where these states are labeled $6\,S_{1/2}$ and $6\,D_{3/2}$. Following the laser excitation, the intermediate $6\,P_{1/2}$ was empty and the high gain on the $6\,D_{3/2}$-$6\,P_{1/2}$ transition allowed superfluorescence (SF) to develop. We observed SF both along and against the direction of the laser excitation pulse. Our working parameters ensured that the SF developed long after the application of the excitation so that parametric amplification processes were not present. We found that, contrary to intuition, SF developed first against the laser direction. We also found that SF was more intense in this backward direction. We also found that SF cascades all the way to the ground state and that quantum beats associated with the hyperfine splitting of the $6\,P_{1/2}$ state were present. In the forward direction radiation appeared simultaneously on both upper and lower transitions. In the presence of a transverse magnetic field we observed time-delayed second harmonic generation.

14. SUBJECT TERMS

DTIC QUALITY INSPECTED 5

superradiance, superfluorescence, nonlinear optics, second harmonic generation, photon echoes

15. NUMBER OF PAGES 6

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

TWO-PHOTON COOPERATIVE CASCADE SUPERFLUORESCENCE

FINAL REPORT

S. R. HARTMANN

10/13/95

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PROBLEM STUDIED

We wanted to study the time development of superfluorescence (SF) in a three-level system which is impulsively excited by way of a two-photon transition. The initial condition is to be one in which the intermediate state is empty while the ground and uppermost states (of the same parity) are populated and are in a coherent superposition. The upper transition is then inverted and SF can be expected if the optical gain is sufficiently high [1]. SF in the backward direction (against the laser propagation direction) is not special and has been studied in great detail [1-6]. In the forward direction it is special as the coherent population transfer it makes to the intermediate state will develop a macroscopic polarization associated with the lower transition. This acts back on the polarization associated with the upper transition and it is this effect we wish to investigate [7, 8].

Since we provide a two-photon excitation we can expect to be able to generate 2nd harmonic (SH) radiation if we apply a transverse magnetic field [9, 10]. We studied the temporal evolution of this SH.

We worked with an atomic vapor of Cs atoms. This choice was dictated by our desire to have a heavy atom so that the inhomogeneous broadening was reduced and coherent radiation processes would dephase slowly. By slow we mean relative to the duration of the two-photon laser excitation pulse which was 10 ps long.

SUMMARY OF MOST IMPORTANT RESULTS

We found that two-photon cooperative cascade superfluorescence, now called yoked superfluorescence, develops as expected [11-13]. It appears simultaneously on the upper and lower optical transitions. It is delayed from the normal SF which develops against the direction of the laser excitation. All these emissions are delayed long after the two-photon laser pulse which sets the initial condition. The SF in the backward direction appears first followed by the yoked SF. On occasion we even observed cascade SF in the backward direction. When present the cascade SF on the lower transition was significantly delayed from the yoked SF.

We made the first unambiguous observation of quantum beats in SF [12]. This was established by the phase locked character they exhibited on the upper and lower transitions.

We made the first observation of time delayed 2nd harmonic generation [14]. Until this experiment 2nd harmonic genreation was always understood to develop simultaneously with the radiation which excited it.

We developed an effective spectral stabilization method for producing stable amplified 10 ps laser pulses at 885 nm [15].

PUBLICATIONS:

1 H. Brownell, B. Gross, X. Lu, S. R. Hartmann, and J. T. Manassah, Coherently inhibited amplification, Laser Phys. **3**, 509 (1993).
2 X. Lu, J. H. Brownell, and S. R. Hartmann, Coherence Inhibition in Cascade Superfluorescence, Laser Phys. **5**, 522 (1995).
3 X. Lu, J. H. Brownell, and S. R. Hartmann, Spectral stabilization of amplified pulses from synchronously-pumped dye laser systems, Opt. Comm. **120**, 295 (1995).
4 J. H. Brownell, X. Lu, and S. R. Hartmann, Yoked Superfluorescence, Phys. Rev. Lett. **75**, No. 16 (1995).
5 J. H. Brownell, X. Lu, and S. R. Hartmann, Time delayed second harmonic generation, Phys. Rev. Lett. **75**, No. 18 (1995).
6 X. Lu, J. H. Brownell, and S. R. Hartmann, submitted for Proceedings of 5th ISSP International Symposium, University of Tokyo, Tokyo, JAPAN, 1995. To appear in a special issue of Progress in Crystal Growth & Characterization of Materials.

PARTICIPATING SCIENTIFIC PERSONNEL

Hayden Brownell, received Ph.D in 1995
Xingmin Lu, received Ph.D. in 1995

DISCUSSION

Theoretical calculations relating to the temporal evolution of cooperative superfluorescence (SF) in a three level system are contained in [16] and in the Ph.D. thesis of X. Lu. Reference [16] treats a pulse propagating on the upper high-gain transition of a three level system prepared by a delta function excitation pulse two-photon resonant with the ground and uppermost state. This analysis allows for effects of large population transfers. It shows that pulse amplification is inhibited by the existence of a coherent superposition between the ground and uppermost state. It also shows that the input pulse on the upper transition generates a copropagating pulse on the lower transition. Lu's thesis deals with a sample similarly two-photon excited but subjected to a noise field which simulates the effect of spontaneous emission. This is the true superfluorescence problem. SF is analyzed both for the case in which a coherent superposition does and does not exist. We find that the coherent superposition inhibits SF.

The two-photon laser pulse required for the excitation of the $6\,D_{3/2}$-$6\,P_{1/2}$ coherent superposition state was at 885 nm. This is a very difficult wavelength to work at when using an amplified synchronously pumped dye laser. This gave us much trouble and was not amenable to conventional stabilization techniques. We finally had to develop a new technique which is published in [15]. The trick was to passively filter the synchronously pumped dye laser output prior to amplifying it. We show the scheme below where SF here stands for spectral filter. DL and PDA are the dye laser and pulse dye amplifiers respectively. G1 and G2 are gratings.

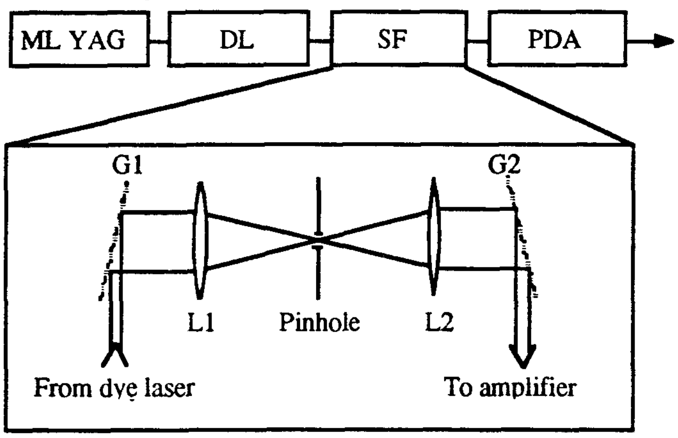

Figure 1. Schematic diagram showing spectral filter developed to increase spectral stability.

With this set up we effectively obtained transformed limited laser pulses 10 ps in duration.

Our spectrally stable pulses made it possible to observe what we initially called cooperative SF. Since superfluorescence is itself cooperative we have decided to use instead the name yoked superfluorescence [12]. Yoked means tied together and implies a slowing down. This is what we observed. We first reported some preliminary results in a laser physics workshop whose proceedings were contained in [13]. A more developed report is provided by reference [12]. We see yoked SF in the forward direction (along the laser excitation pulse). By this we mean the simultaneous existence of superradiance on both the upper $6\,D_{3/2}$-$6\,P_{1/2}$ and lower $6\,P_{1/2}$-$6\,S_{1/2}$ transitions. This SF was delayed from the 10 ps excitation pulse by more that 100 ps. We emphasize that these signals appeared at the same time. They were yoked, as it were. In

the backward direction we also see SF on both the upper 6 $D_{3/2}$-6 $P_{1/2}$ and lower 6 $P_{1/2}$-6 $S_{1/2}$ transitions, but these emissions were not simultaneous. This is properly called cascade SF. First SF appears on the upper transition; when this terminates the resulting population transfer can give rise to an inversion on the lower transition. This can cause SF to develop on the lower transition. Our observation of cascade SF terminating on the lower transition is a first.

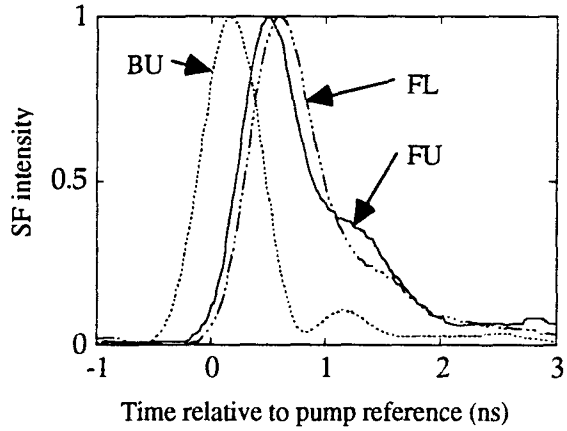

Figure 2 Normalized SF responses for backward upper (BU), forward upper (FU) and forward lower (FL) transitions in Cs at 150 C pumped with 5×10^{10} photons. The BL signal was too small to detect. The yoked character in the forward direction is clearly displayed.

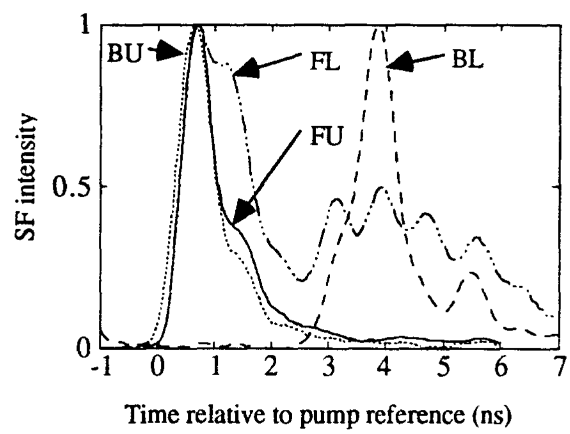

Figure 2 Normalized SF responses for BU, FU, BL,and FL transitions in Cs at 110 C pumped with 1×10^{11} photons. Notice the quantum beats. Here we see cascade SF terminating in the ground state.

We see then the upper leg of the cascade SF propagating in the backward direction followed by yoked SF propagating in the forward direction followed in turn by the lower leg of the cascade SF propagating in the backward direction. This is the major result of our experiment.

As a bonus we also observe quantum beats. This is the first definite quantum beat observation in SF as it appears in both legs of the cascade and yoked emissions and is phase locked. It is documented in Brownell's Ph.D. thesis as well as in [12].

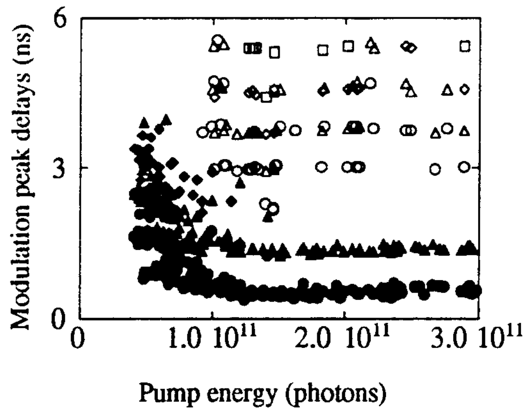

Figure 4 Quantum beat display. Modulation peak delays (referred to pump) vs pump energy in Cs at 110 C for the BU (solid shape) and BL (open shape) transitions. The sequence of shapes (circle, triangle, diamond, square) corresponds to the sequence of modulation peaks identified in a single response.

SF is inherently noisy and the statistics it displays are inherently interesting. Some data is contained in Brownell's thesis but more data and analysis is needed to properly appreciate it.

In the presence of a transverse magnetic field 2nd harmonic (SH) generation is produced [9]. We carefully time resolved this emission and found that it was delayed from the excitation pulse that generated it. This experiment is described in [14]. The reason it is delayed is understood by noting that the superposition state generated by the two-photon resonant excitation is the same independent of whether or not the magnetic field is present. But in the absence of a magnetic field, 2nd harmonic generation is forbidden in an atomic vapor. Thus no 2nd harmonic is produced by the action of the laser pulse. The effect of the magnetic field is to induce transitions which induce a radiating electric quadrupole moment. This takes time and in our experiment corresponded to several hundred ps. Our observation of delayed 2nd harmonic generation is the first of its kind.

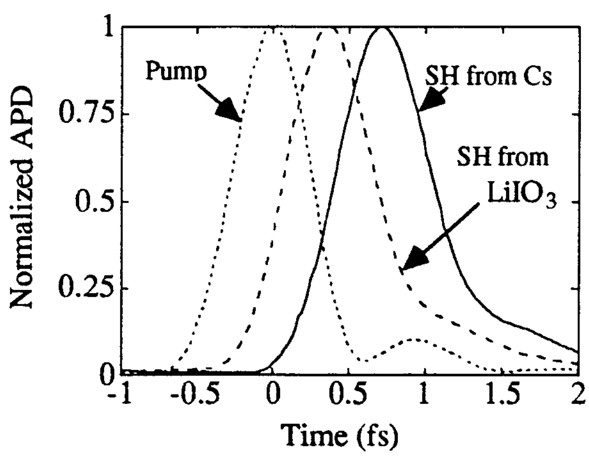

Figure 5 Temporal profiles of (1) SH from Cs in a 570 Gauss field, (2) laser pump referenced to 442 nm by detecting $LiIO_3$ SH, and (3) laser pump detected at 885 nm.

In Fig. 5 the SH from $LiIO_3$ was used to reference the pump position since the avalanche detectors we used had a response time that was wavelength dependent.

Not yet submitted for publication is the result of our photon echo experiments working on the $6\,P_{1/2}$-$6\,S_{1/2}$ transition. We observe quantum beats associated with both the 1.168 GHz and 9.193 GHz hyperfine splittings of the excited and ground states. We have performed an analysis which applies to all the alkalies. This analysis incorporates Billiard Ball diagrams in such a way as to provide deeper insight into the echo modulation process.

REPORT OF INVENTIONS

None.

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